Influence of uncertainty in atmospheric state on the accuracy of gas analysis by the differential absorption lidar method in 9–11 and 4.6–5.6 μm spectral regions

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The general equation is derived for the error due to approximate description of the atmospheric state based on the statistical model of the atmosphere and some atmospheric parameters measured in lidar sounding. It is shown that the measurement of additional atmospheric parameters can significantly reduce the uncertainty in concentration of the gas under study. The computer code has been developed to search for frequency pairs among CO_2 laser lines, which give minimum error of measurements along a medium-length horizontal path with a topographic target as a retroreflector. The calculations show that some previous experiments on remote measurement of atmospheric gas concentrations were conducted at frequencies that did not provide for the minimal atmospheric error.

Introduction

The CO_2-laser based lidars are now extensively used in remote gas analysis of the atmosphere by the differential absorption lidar (DIAL) method. The lidars operate in the 9–11 μm spectral region of the fundamental laser lines and in the 4.6–5.6 μm region of sum frequencies. The concentrations of NH_3, O_3, H_2O, SO_2, C_2H_4, NO, CO, and other gases were determined with the use of CO_2-laser based lidars.[1–9]

The accuracy of atmospheric gas concentration measurements depends heavily on the proper choice of working frequencies. The general algorithm for selection of optimal spectral channels for gas analysis was proposed in Refs. 10 and 11. It is based on calculation of the informative distance in the space of states of a gaseous object under study. To implement this algorithm, one needs to know the absorption coefficient of the gas under study, atmospheric transmission spectrum, and the variance of the noise component of a lidar return signal. The noise in a lidar return signal can be caused by an instability of the laser radiation frequency, of its mode structure, and direction, as well as by the detector and amplifier noise, speckle structure and spots of a topographic target, scintillation at scattering on aerosols, and turbulent fluctuations of optical characteristics of the atmosphere. The variance of fluctuations of this type can be reduced significantly by signal averaging over a large number of recorded pulses.[5,12]

There are some other sources of errors impairing the measurement accuracy similarly to the noise in return signals. However, their contribution cannot be reduced by averaging over a series of pulses. These sources include inaccurate knowledge of the absorption coefficient of the gas under study and spectral dependence of the coefficients of aerosol scattering and reflection from a target. The role of these factors was assessed in Refs. 13–15. This type of errors also includes absorption by interfering gases, whose content in the atmosphere is not accurately known. This problem was discussed in Refs. 5, 13, and 16–18, where it was concluded that the practical limit of the accuracy of concentration measurements in the 9–11 μm spectral region is determined by variations of the atmospheric transmittance during the time of averaging.

The influence of uncertainties in the atmospheric parameters on the accuracy of gas concentration measurements in the 9–11 μm region was studied in Ref. 18, in which it was shown that an incorrect consideration of the absorption by interfering gases can result in the errors in the concentration of the gaseous constituent under study of hundreds and even thousands percents as high. In this connection, it is recommended to take into account specific meteorological conditions to optimize the measurements.

The nature of the discussed atmospheric error is the following. When estimating the concentration from a signal value, one needs to know the temperature, on which the absorption depends, and to subtract the contribution coming from interfering constituents to the total absorption. In other words, one needs either to measure in parallel all the atmospheric parameters, the measured concentration depends on, or to use some model of the atmosphere, which includes typical mean values of the temperature and concentrations. In the latter case, the error is introduced, whose value is related to the range of possible variations of atmospheric parameters about their mean values. However, even in the case of parallel measurements, the natural variability of the atmosphere in space and time restricts the accuracy of measuring the parameters of an atmospheric path during the period of sounding. It is a common practice to complement measurements with model data. For example, in Ref. 19 lidar measurements of humidity used data on the vertical temperature profile acquired with a radiosonde, whereas the atmospheric pressure was taken from the model of the standard atmosphere.

In Ref. 18 it was assumed, in fact, that the measurement error is determined by the total content of the interfering constituents in the atmosphere. In estimating the error, their concentrations are borrowed from the corresponding statistical models of the atmosphere. The error calculated here is determined by the accuracy of setting the temperature and humidity, as well as the variability of the content of interfering constituents, which is low in some cases as compared to their mean

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concentrations. Various conditions of measurements (horizontal paths, slant paths, backscatter method, use of topographic targets) are considered. In some cases, the error caused by uncertainties in the atmospheric parameters significantly limits the measurement accuracy. However, if the data of field measurements of some atmospheric parameters, on which the transmittance depends significantly, are used along with the atmospheric model, then the accuracy of concentration measurements can be markedly improved.

The equations obtained allow one to find the optimal frequency pairs, for which the error due to uncertainty in the atmospheric state is minimal, and to evaluate the need for invoking additional information, for example, surface values of the atmospheric temperature and humidity.

1. Theory

The "on" and "off" frequencies with the correspondingly strong and weak atmospheric absorption will be considered to be close enough, so that the frequency dependence of the scattering and reflection coefficients can be neglected.

If reflection from a topographic target is used to obtain a return signal, then the concentration averaged over the sounding path can be estimated as

$$U_t = \frac{1}{2} \ln \frac{P'(R)}{P(R)} = \int\limits_0^R k(x)\, \mathrm{d}x\,, \tag{1}$$

where R is the path length; P and P' are return signal power at the "on" and "off" lines. If the spectral width of a laser line is much less than the typical width of an absorption line of the gas under study, then k is the difference between the volume absorption coefficients at the "on" and "off" frequencies. Otherwise, as was shown in Ref. 20, k is the difference between the atmospheric absorption coefficients, which are averaged over spectral profiles of separate laser lines with the weight depending on the atmospheric transmittance along the sounding path. If the lidar is operated in the backscatter mode, in which the portion of light scattered back by the atmosphere is received, the local concentration can be found from the equation

$$U_r = \frac{1}{2} \ln \frac{P'(R)\, P(R+\Delta R)}{P(R) P'(R+\Delta R)} = k(R)\, \Delta R. \tag{2}$$

In both of these cases, estimation of the concentration is reduced to analysis of the difference between the absorption coefficients at the sounding frequencies. Assuming, in what follows, that laser lines are monochromatic, let us present the difference between the absorption coefficients in the following form:

$$k = \sum_{j=2}^{M} \sigma_j\, N_j + k_{H_2O}(N_1), \tag{3}$$

where M is the number of constituents contributing to absorption at the sounding frequencies; σ_j is the difference between the absorption cross sections at the "on" and "off" frequencies of the jth constituent of a gas mixture; the cross sections depend mostly on pressure and temperature; N_j is the concentration of the jth constituent of a gas mixture; N_1 is the water vapor concentration; k_{H_2O} is the difference between the water vapor absorption coefficients accounting for the selective and continual absorption. To determine the concentration of the gas under study, an atmospheric model will be used in combination with the measurement data on some atmospheric parameters, for example, temperature and humidity at one or several altitudes. Let us estimate the error in this case. For gases with the linear concentration dependence of the absorption coefficient, it is easy to derive the equation for the local or path-averaged concentration from Eqs. (1)–(3). We present here only the equation for the case of reflection from a topographic target. If not measured the atmospheric parameters are taken from an atmospheric model, then the concentration of the ith gas ($i \neq 1$) averaged over the sounding path with the weight equal to the cross section difference is equal to

$$N_i^* = \left\{ U_t(R) - \int\limits_0^R {\sum_{j=2}^{M}}' \sigma_j^*(x)\, N_j^*(x) + k^*_{H_2O}(x)\, \mathrm{d}x \right\} \times \\ \times \left[\int\limits_0^R \sigma_i^*(x)\, \mathrm{d}x \right]^{-1}, \tag{4}$$

where the asterisk denotes that the corresponding parameter is determined with the use of mean temperature and mean concentration borrowed from an atmospheric model. Primed sum signs mean that the term with $j = i$ is excluded from the sum.

Possible variations of the measured concentration due to deviation of the actual atmospheric parameters from the model ones, as well as due to signal fluctuations can be presented in the form

$$\Delta N_i = (\Delta U_t - \Delta S_a) / \int\limits_0^R \sigma_i^*(x)\, \mathrm{d}x. \tag{5}$$

Here ΔU_t is the fluctuation of the parameter U_t due to the power fluctuations of the received signals P and P', and ΔS_a is the variation of the difference between the optical thicknesses of the path at the "on" and "off" frequencies:

$$\Delta S_a = \int\limits_0^R {\sum_{j=0}^{M}}' \sigma_j^*(x)\, \Delta N_j(x)\, \mathrm{d}x, \tag{6}$$

where for brevity the following designations are used:

$$\sigma_0 = \partial k / \partial T, \quad \sigma_1 = \partial k_{H_2O} / \partial N_1;$$

$$\Delta N_0 = \Delta T = T - \overline{\overline{T}}, \quad \Delta N_j = N_j - \overline{\overline{N_j}}. \tag{7}$$

The doubled overbar denotes averaging over variations of atmospheric parameters accepted by the atmospheric model in use (in fact, for a many-year period). In the case of the backscatter mode of sounding, the local concentration and its fluctuations are described by equations similar to Eqs. (4)–(6) with the integrals replaced by the products of the local values of integrands by ΔR and U_t replaced by U_r.

Taking into account that the noise in a signal does not correlate with the variations of atmospheric temperature and composition, we obtain from Eq. (5) that

$$\langle N_i \rangle = N_i^* + \langle \Delta N_i \rangle, \ \langle \Delta N_i \rangle = -\langle \Delta S_a \rangle \Big/ \int\limits_0^R \sigma_i^*(x) \mathrm{d}x, \tag{8}$$

$$D(\Delta N_i) = \langle \Delta N_i^2 \rangle - \langle \Delta N_i \rangle^2 = \\ = \left(\langle \Delta U_t^2 \rangle + D(\Delta S_a)\right) \Big/ \left(\int\limits_0^R \sigma_i^*(x)\,\mathrm{d}x \right)^2, \qquad (9)$$

where $D(q)$ is the variance of the random variable q.

With no information available on the atmospheric temperature and concentrations of the absorbing atmospheric constituents at the time of measurement, the unconditional averaging takes place, in which $\langle \Delta S_a \rangle$ is zero, and $D\langle \Delta S_a \rangle$ is determined by the average values and covariation functions of temperature and concentrations from the statistical model of the atmosphere. However, to calculate the conditional means (provided that some atmospheric parameters are known), one needs the conditional probability of ΔS_a, i.e., the joint function of the probability of temperature and concentrations at different altitudes. Since such information is absent now, we take that this probability density function can be approximated by a multidimensional Gauss distribution with the mean values and second moments coinciding with the corresponding parameters from the statistical model of the atmosphere. Then the joint distribution function of the atmospheric parameters and signal variations ΔS_a is also a multidimensional Gauss distribution function because of the ΔS_a linear dependence on the variations of temperature and concentrations.

Deviations of the measured atmospheric parameters from their model means are denoted as Z_α, where $\alpha = 1, 2, \ldots P$, and P is the number of the measured atmospheric parameters. For example, at $Đ = 3$ $Z_1 = \Delta T(0)$, $Z_2 = \Delta T(1)$, and $Z_3 = \Delta N_1(1)$ are the deviations of temperature and water vapor concentration from their many-year means at the sounding altitudes of 0 and 1 km.

In Eqs. (8) and (9) we first average over variations of the atmospheric parameters provided that P parameters Z_α take some particular values. The result of such averaging depends on the second moments according to the statistical model of the atmosphere and particular values of Z_α. After one more averaging over the possible spread of the measurement results on Z_α, we have

$$\langle \Delta S_a \rangle = \sum_{\alpha=1}^{P} y_\alpha \, \overline{Z_\alpha}\,, \qquad (10)$$

$$D(\Delta S_a) = \langle \Delta S_a^2 \rangle - \langle \Delta S_a \rangle^2 = \overline{\overline{\Delta S_a^2}} - \sum_{\alpha=1}^{P} y_\alpha B_\alpha + \\ + \sum_{\alpha,\beta=1}^{P} y_\alpha y_\beta \, \overline{\delta Z_\alpha \, \delta Z_\beta}\,, \qquad (11)$$

where the overbar denotes averaging over the array of the measurement results on Z_α;

$$\delta Z_\alpha = Z_\alpha - \overline{Z_\alpha}\,; \qquad (12)$$

$$\overline{\overline{\Delta S_a^2}} = \int\limits_0^R \int\limits_0^R \mathrm{d}x_1 \, \mathrm{d}x_2 \sum_{j,k=0}^{M}{}' \sigma_j(x_1) \, \sigma_k(x_2) \, \overline{\overline{\Delta N_j(x_1) \, \Delta N_k(x_2)}} \qquad (13)$$

$$y_\alpha = \sum_{\beta=1}^{P} (K^{-1})_{\alpha\beta} \, B_\beta\,; \qquad (14)$$

$$B_\alpha = \overline{\overline{\Delta S_a \, Z_\alpha}} = \int\limits_0^R \sum_{j=0}^{M}{}' \sigma_j^*(x) \, \overline{\overline{\Delta N_j(x) \, Z_\alpha}} \, \mathrm{d}x; \qquad (15)$$

K^{-1} is the matrix inverse to K:

$$K_{\alpha\beta} = \overline{\overline{Z_\alpha \, Z_\beta}}. \qquad (16)$$

The matrix K is calculated using a statistical model of the atmosphere, and $\overline{\overline{\Delta S_a^2}}$ and B_α are calculated with the use of a spectroscopic database and a statistical model of the atmosphere. These values are then used to determine y_α and then $\langle \Delta S_a \rangle$ and $D(\Delta S_a)$. At $Đ = 0$ (unconditional averaging) all the sums in Eqs. (10) and (11) are believed zero. This method can be considered as an extended method of invoking additional information, which was used in Refs. 21 and 22 in calculating the atmospheric transmittance.

Equations (10) and (11) are valid for any paths, including horizontal ones. However, in the case of homogeneous near-ground horizontal paths, the set of the atmospheric parameters describing the average state of the atmosphere is worth including directly into the measured parameters instead of the model ones. Let us present equations for the mean value and the variance ΔS_a for the case of measurements along homogeneous near-ground horizontal paths with a topographic target in an explicit form. Assume that we know only temperature T and relative humidity φ of the atmosphere accurate to some error. Then, expressing the variation of the concentration of a gas under study with respect to the new average state of the atmosphere and then making the above-mentioned averaging, we obtain

$$\langle \Delta S_a \rangle = 0; \qquad (17)$$

$$D(\Delta N_i) = \left(\langle \Delta U_t^2 \rangle + R^2 \left\{ \overline{\delta^2 T} \left[\sigma_0 + \sigma_1 \frac{\overline{N}_1}{\overline{T}} \left(\frac{T_e}{\overline{T}} - 1 \right) \right]^2 + \\ + \overline{\delta^2 \varphi} \; \sigma_1^2 \, \overline{N}_1^2 \, \varphi^{-2} + \sum_{j=2}^{M}{}' \sigma_j^2 \, \overline{\overline{\Delta N_j^2}} \right\} \right) \Big/ R^2 \sigma_i^2. \qquad (18)$$

In Eq. (18) the differences between the cross sections σ_j are determined through the new set of parameters describing the average state of the atmosphere; T_e is the energy of vaporization of one water molecule, in K, $T_e \approx 5600$ K; $\overline{N}_1$ is the mean concentration of water molecules calculated from the mean air temperature $\overline{T}$ and humidity $\overline{\varphi}$. At a direct detection of a lidar return signal in the infrared spectral region, the error due to the noise in the recording system is caused by the dark current of a photoresistor and fluctuations of the gain factor of an amplifier. These factors as well as return signal fluctuations due to speckles and atmospheric turbulence are taken into account according to Refs. 23 and 24. Thus we have the following equation:

$$\langle \delta^2 U_i \rangle = \left(\langle \delta^2 A \rangle + 4\gamma^2 + \frac{W^2}{P^2} + \frac{W^2}{P'^2} \right) \Big/ N, \qquad (19)$$

where γ is the relative rms fluctuation of the amplification coefficient; in typical cases the order of magnitude of this value is equal to 0.01; N is the number of pulse pairs to be averaged; $W = NEP(2\Delta f)^{1/2}$, NEP is the noise equivalent power, Δf is the amplifier pass band; $\langle \delta^2 A \rangle$ is the contribution from speckles and atmospheric turbulence to the signal variance.

2. Calculated results

To estimate numerically the error due to uncertainty in the atmospheric state when measuring gas concentration along inhomogeneous paths, the computer program was developed, which includes atmospheric and optical blocks. The atmospheric block is based on the statistical model of the atmosphere[25] and the data from Ref. 26: altitude distributions of variances, altitude covariations of temperature, humidity, ozone, and other gases obtained in many-year observations. Besides, the data of Minsk Observatory on the covariations of atmospheric temperature and humidity were used. Covariations for which we had no data were assumed zero. According to the atmospheric model used, different components of the atmosphere, except for temperature and humidity, do not correlate with each other. The atmospheric block is used for calculation of the mean values of atmospheric parameters and their possible variations along the sounding paths.

The optical block is used for calculation of the absorption coefficient and transmittance of the atmosphere from the temperature, humidity, and concentration distributions obtained using the atmospheric block. The HITRAN–96 database[27] provides the initial information for the optical block. According to this database, eight substances (H_2O, CO_2, O_3, N_2O, CH_4, SO_2, NO_2, and NH_3) contribute to absorption in the 9–11 μm wavelength region, and ten substances (all the above-listed plus NO and CO) contribute to absorption in the 4.6–5.6 μm region. Besides, the data on the absorption coefficients of other gases at the laser frequencies were used.[8] The absorption coefficients of the gases included in HITRAN–96 were calculated taking into account all the lines with centers lying no farther than 20 cm^{-1} from the frequency under consideration. Spectral lines were assumed to have the Voigt profile. The water vapor absorption coefficient was calculated as a sum of selective and continual absorption.

Let us give some examples of calculations by the developed program. To estimate the expected error of long-path measurements in the case of a slant path, Eqs. (9) and (11)–(16) should be used. In the case of the backscatter mode, the integrals in Eqs. (13) and (15) should be replaced by the products of the local values of the integrands at the sounding point by the scale of the device's spatial resolution ΔR. Since we are interested, first of all, in the atmospheric component of the error, the signal fluctuation $\langle \Delta U^2 \rangle$ in Eq. (9) is assumed zero. In practice it can be significantly reduced by averaging over a large number of pulses.

Figure 1 shows the vertical profiles of the non-conditional and conditional errors of SO_2 concentration measured from the ground in the backscatter mode at the frequencies $\nu_{\text{on}}/\nu_{\text{off}} = 1077.023/1044.591\,\text{cm}^{-1}$. For calculation of the non-conditional error, all the atmospheric parameters needed for estimating the concentration of the substance sounded are borrowed from the model, whereas for calculation of the conditional error the measurement data on some of the atmospheric parameters at the time of sounding are additionally used. One can see that the use of data of surface measurements of temperature and humidity only slightly decreases the atmospheric component of the error. This is because the uncertainty in the O_3 content, rather than in temperature and humidity, contributes mostly to the error at the frequencies used. Therefore, to decrease the error, it is worth measuring first, the ozone profile and then using it in signal processing for determination of the SO_2 concentration. This procedure allows the atmospheric component of the error to be decreased down to 0.1–0.2 ppm at altitudes not exceeding the vertical scale of correlation of the atmospheric parameters.

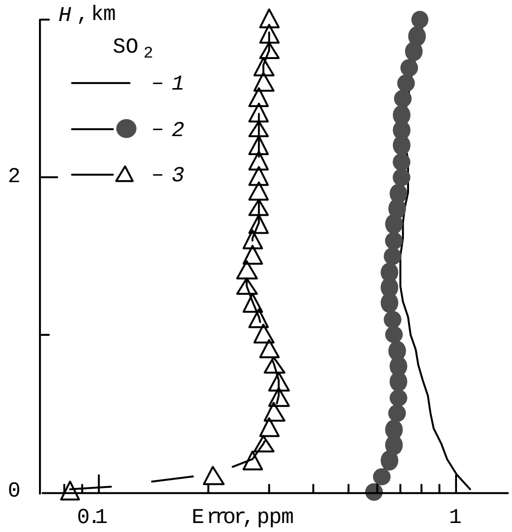

Fig. 1. Vertical distributions of the error of SO_2 measurement in the backscatter mode: non-conditional error (*1*); conditional error (*2*) (surface measurements of temperature and humidity were used); the same as curve *2* plus the use of the ozone profiles measured accurate to 1 ppb (*3*).

Figure 2 shows the calculated errors of measurement of the C_6H_6 concentration. The calculations simulate airborne measurements in the mode of recording lidar returns from the surface. Mayer et al.[8] recommended that the C_6H_6 concentration be measured at the frequencies 1039.393 (on) and 1078.516 cm^{-1} (off). Figure 2 shows the measurement error in the C_6H_6 concentration averaged over the 0 – H km atmospheric layer as a function of the flight altitude H (for measurements at the recommended frequencies). As is seen, measurement of the near-ground values of temperature, humidity, and ozone concentration significantly reduces the expected error in the benzene vapor concentration (in this case, down to the value of about 0.05 MPC). The MPC (maximum permissible concentration) for C_6H_6 vapor is equal to 5 mg/m^3, i.e., about 2 ppm under standard conditions. Measurements from low altitudes are characterized by higher intensity than those from high altitudes because of correlation of the atmospheric parameters at different altitudes.

The code described above made the basis for the Optipa program intended for seeking of optimal, i.e., corresponding to minimal errors, conditions of horizontal measurements with a topographic target as a retroreflector. The automatic selection of the optimal measurement conditions based on minimization of possible errors was proposed, for example, in Refs. 18 and 28. Of special note is the versatile software package described in Ref. 28, which allows one to optimize the conditions of H_2O and O_3 sounding by various lidars operating in the UV and IR regions. The Optipa program allows optimization of concentration measurements for various atmospheric gases to be performed in the spectral regions of the basic and sum frequencies of the CO_2 laser. The input data for the program are laser frequencies and energies of laser pulses, as well as mean concentrations of the atmospheric gases, (including those under study), atmospheric pressure, temperature, relative humidity, and their rms errors. Then the following parameters are set: maximum frequency separation between the laser lines, path length, reflection coefficient of a target, aperture of the receiving telescope, optical path transmittance, quantum efficiency of the detector, relative amplification of the error γ, noise equivalent power W. All the parameters are entered in the dialog regime.

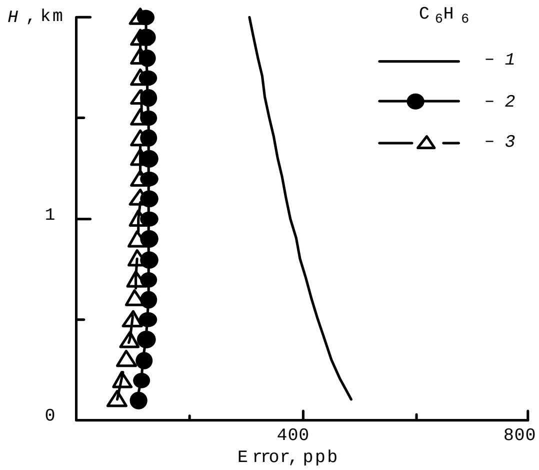

Fig. 2. Errors of determination of the benzene concentration averaged over the whole thickness of the atmospheric layer for measurements from an aircraft flying at the altitude *H*; absolute error (*1*); conditional errors (*2* and *3*) (curve *2* is obtained for the case of temperature and humidity measured at $H = 0$; curve *3* was obtained with temperature, humidity, and ozone concentration measured at $H= 0$).

Table 1 gives the error ΔN (in ppb) in the mean concentration due to uncertainties in atmospheric parameters for measurements along horizontal paths with topographic targets. The error was calculated by Eq. (18) for both of the frequencies recommended in the literature and those found with the Optipa program. In calculations the mid-latitudinal summer model of the atmosphere was used. The mean temperature $T = 293$ K and the mean humidity of 70% were assumed known at the period of measurements accurate to 1 K and 1%, respectively.

Table 1. Concentration errors due to uncertainty in the state of the atmosphere

Gas	Ref.	ν_{on}	ν_{off}	$\Delta\nu$, cm^{-1}	ΔN, ppb	Err, %	Source of error
NH_3	Ref. 2	9R(30)	9R(26)	2.34	$9.4 \cdot 10^{-2}$	0.28	T, CO_2, H_2O
	*	9R(30)	9R(26)	2.34(5)	$9.4 \cdot 10^{-2}$	0.28	T, CO_2, H_2O
ÑO	Ref. 2	10P(32)	10P(30)	1.93	0.152	$9.4 \cdot 10^{-2}$	CO_2, T, H_2O
	Ref. 5	2·9R(18)	2·9R(20)	2.58	4.0	5.2	H_2O, T
	*	2·9R(30)	2·9R(34)	446(5)	0.53	0.56	N_2O, T
NO	Ref. 5	2·10P(24)	2·10P(14)	17.9	42.9	4.2	H_2O, T
	*	2·10P(24)	2·10P(14)	17.9(18)	42.9	4.2	H_2O, T
	*	2·10P(24)	2·10P(26)	3.72(5)	48.6	5.0	H_2O, T
SO_2	Ref. 3	9R(34)	9R(24)	5.78	924	0.76	T, CO_2, H_2O
	*	9R(26)	9R(22)	2.44(6)	201	0.24	CO_2, T, NH_3
O_3	Ref. 6	9P(14)	9P(24)	9.03′	0.57	0.28	NH_3, CO_2, T
	*	9P(14)	9P(22)	7.17(10)	0.19	$8.8 \cdot 10^{-2}$	H_2O, CO_2
H_2O	Ref. 9	10R(20)	10R(18)	1.31	$3.79 \cdot 10^{5}$	10.8	T
	*	10R(30)	10R(28)	1.18(5)	$1.76 \cdot 10^{5}$	$1.2 \cdot 10^{-2}$	T, CO_2
	*	2·10P(22)	2·10P(20)	1.81(5)	590	0.50	T, NO

* Our results.

In the table, $\Delta\nu$ means the difference between the frequencies ν_{on} and ν_{off}; the maximum permissible difference $\nu_{on} - \nu_{off}$ used in the search for optimal pairs is given in the parenthesis; Err is the error of the ratio between the signals at ν_{on} and ν_{off} with the target at a 1-km distance. The last column gives the main factors leading to errors at the given frequency pair: gases whose absorption spectrum overlaps that of the gas under study and temperature (T).

The frequency pairs found by us for NH_3 and NO coincide with those given in the literature. For other gases we found the frequency pairs, different from that already known, at which the error due to uncertainty in the state of the atmosphere is less than that for the latter. For the NO molecule, we propose the pair of lines with the error slightly exceeding that for the pair known in the literature. However, the proposed lines are closer to each other, what is important for elimination of the spectral dependence of the reflection and scattering coefficients. It is noting the decrease of the atmospheric error in measurements of the water vapor content at the sum frequencies of the CO_2 laser as compared with the measurements in the 9–11 μm region. The results of the refined calculation of optimal pairs, taking into account the noise of the recording system and fluctuations of the return signal and atmospheric turbulence, will be published in the next paper.

Conclusion

One of the potential sources of errors arising in remote measurement of concentrations of atmospheric gases is an incomplete knowledge of the atmospheric composition and temperature during the measurements. In this paper we have derived the general equation for the error due to approximate description of the atmospheric state based on the statistical model of the atmosphere using data on some atmospheric parameters measured at the time of lidar sounding. To estimate numerically this error, the computer program involving the atmospheric and optical blocks has been developed. The atmospheric block is based on the statistical model of the atmosphere[25,26] including altitude distributions of the mean concentrations and temperature, their variances and altitude covariations. The optical block is intended for calculation of the absorption coefficient and atmospheric transmittance from the distributions of temperature, pressure, and concentrations determined in the atmospheric block. The initial information for the optical block is taken from the HITRAN–96 database.[27] The continuum water vapor absorption is taken into account as well.

The calculations show that the use of additional data on the atmospheric parameters can reduce significantly the uncertainty in the concentration of the gas under study. The code described above has made the basis for the specialized program intended for seeking of pairs of CO_2 laser lines corresponding to minimal errors at horizontal sounding with a topographic target as a retroreflector.

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